FORM PTO-13: (REV 11-2000)	90 (Modified) U.S. DEPARTMENT	FOF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER				
		TO THE UNITED STATES	212638US0PCT				
DESIGNATED/ELECTED OFFICE (DO/EO/US) U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR							
		NG UNDER 35 U.S.C. 371	09/913717				
	TIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED				
	PCT/JP00/01088 NVENTION	February 25, 2000	February 25, 1999 (earliest)				
		INC MATERIAL VIRRATION DAM	PING PART AND COMPOSITION FOR				
COPOLY	MER TOTAL	ING WATERIAL, VIDRATION-DAME	FING PART AND COMPOSITION FOR				
	T(S) FOR DO/EO/US URA Hiroki et al.						
I VITAL ALVA	UKA IIII OKI EL AI.						
A policant l	ith anti-it- to the United Cte						
		ttes Designated/Elected Office (DO/EO/US) th					
1.		tems concerning a filing under 35 U.S.C. 371.					
2.		UENT submission of items concerning a filing					
3. ⊠	This is an express request to beg (6), (9) and (24) indicated below	in national examination procedures (35 U.S.C.	3. 371(f)). The submission must include itens (5),				
4. 🛛		expiration of 19 months from the priority date	(Article 31)				
5. ⊠		lication as filed (35 U.S.C. 371 (c) (2))	(Afficie 51).				
_		tired only if not communicated by the Internat	tional Bureau)				
71000		d by the International Bureau.	nonal Bulcau).				
windows		application was filed in the United States Recei	iving Office (RO/US)				
		of the International Application as filed (35 U					
<u> </u>	a. ⊠ is attached hereto.	or me meeting ripphounten as med (55 C	.5.0. 571(0)(2)).				
		bmitted under 35 U.S.C. 154(d)(4).					
7. 🛛		e International Application under PCT Article	19 (35 H S C 371 (c)(3))				
jimate is. E		uired only if not communicated by the Interna					
*** <u>*</u>		ed by the International Bureau.	Monar Barcaay.				
		owever, the time limit for making such amendn	nents has NOT expired				
	d. 🛮 have not been made and will not be made.						
18. □	An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).						
9. ⊠	An oath or declaration of the inventor(s) (35 U.S C. 371 (c)(4)).						
₩. □	An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).						
11.	A copy of the International Preliminary Examination Report (PCT/IPEA/409).						
12.	A copy of the International Search Report (PCT/ISA/210).						
Items 1	3 to 20 below concern document	·					
13.		ement under 37 CFR 1.97 and 1.98.					
14.		ording. A separate cover sheet in compliance v	with 37 CFR 3.28 and 3.31 is included				
15.	A FIRST preliminary amendmen		and sist is moraded.				
16. 🗆	A SECOND or SUBSEQUENT						
17. 🗆	A substitute specification.						
18.	A change of power of attorney an	nd/or address letter.					
19. \square	A computer-readable form of the	sequence listing in accordance with PCT Rule	e 13ter.2 and 35 U.S.C. 1.821 - 1.825				
20.		nternational application under 35 U.S.C. 154(d					
21.	A second copy of the English lan	guage translation of the international application	on under 35 U.S.C. 154(d)(4).				
22.	Certificate of Mailing by Express	Mail					
23.	Other items or information:						
	Notice for Consideration of Doc	cuments Cited in International Search Repo	ort/Notice of Priority				
	PCT/IB/304/Drawings (1 sheet)	/PCT/IB/308					

U.S. A	PPLICA		<i>o</i> <u> </u>	1 3 7	1 7	PCT/.	, APPLICAT JP00/010 ;		1 O.		ATTORNEY'S 21263		KET NUMBER OPCT
24.	C.							CAI			TO USE ONLY		
BASI	C NAT	IONA	L FEE	(37 CFR	2 1.492 (a) (1) -	(5)):	•			CA	LCULATION	5 P	TO USE ONLY
	Neith intern	er inter ational	mational search:	l prelimin fee (37 C	ary examinatio FR 1.445(a)(2)	n fee (37 CFR 1.482) paid to USPTO I by the EPO or JPO.			\$1000.00				
×	Intern	ational	l prelimi	narv exar	mination fee (3'	7 CFR 1.482) not paid pared by the EPO or J	1 to		\$860.00				
	Intern but in	ational ternatio	l prelimi onal sea	nary exar rch fee (3	nination fee (3' 7 CFR 1.445(a	7 CFR 1.482) not paid (2)) paid to USPTO .	i to USPT	\cap	\$710.00				
	but all	claim	s did no	t satisfy p	provisions of PO	7 CFR 1.482) paid to CT Article 33(1)-(4).			\$690.00				
	Intern and al	ational l claim	is satisfi	ed provis	ions of PCT Ar	7 CFR 1.482) paid to ticle 33(1)-(4)			\$100.00			1	
L						ATE BASIC F	EE AM	OUN	NT =		\$860.00		
month	s from	\$130.0 the ear	liest clai	imed prio	rity date (37 C	· · · · · · · · · · · · · · · · · · ·		20	□ 30		\$0.00		
	AIMS				R FILED	NUMBER EX	TRA	┼	RATE			-	
Total c				5	- 20 =	0		х	\$18.00		\$0.00		
	ependent claims 2 - 3 = 0 x \$80.00					\$0.00							
Multiple Dependent Claims (check if applicable). TOTAL OF ABOVE CALCULATIONS = \$860.00													
re	duced	by 1/2.									\$0.00		
SUBTOTAL = \$860.00						·							
Processing fee of \$130.00 for furnishing the English translation later than 20 30 4 4 50.00 20 4 50.00													
TOTAL NATIONAL FEE						E =		\$860.00					
Eee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).							\$0.00						
						TOTAL FEES	ENCL	OSE	E D =		\$860.00		
										Amou	unt to be: efunded	\$	
							(charged	\$				
a.	a. A check in the amount of to cover the above fees is enclosed.												
<u></u>													
c.	N7 en a												
d.	is displaced copy of this sheet is chelosed.												
NOTE:	NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.												
						e the application to	penumg s	tatus.					
SEND.	ALL C	ORRE.	SPOND	ENCE TO	J:		1	~ \mathre{\pi}	pelleur	المال	AM E. BEA	UMc)N
								218	NATÜRE	STRA	TION NUM	IBE F	1 30,330
								Nor) man F. Ol	hlon			
			11	11 1	[]			NAN		01011	· · · · · · · · · · · · · · · · · · ·		
			H					$\frac{24,6}{8EG}$	ISTRATIO	NI NII II	MDED		
				22	850			NEC.	I ,		VIDEK		
	÷							DAT	re/	17/	1001		
									-				:

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

:

HIROKI NAKAMURA ET AL

: ATTN: APPLICATION DIVISION

SERIAL NO: NEW U.S. PCT APPLN

(Based on PCT/JP00/01088)

FILED: HEREWITH

FOR: COPOLYMER, VIBRATION-

DAMPING MATERIAL, VIBRATION-DAMPING PART AND COMPOSITION FOR

COPOLYMER

PRELIMINARY AMENDMENT

ASSISTANT COMMISSIONER FOR PATENTS WASHINGTON, D.C. 20231

SIR:

Prior to examination on the merits, please amend the above-identified application as follows.

IN THE CLAIMS

Please cancel Claim 2.

Please amend the claims as shown on the marked-up copy following this amendment to read as follows.

1. (Amended) A copolymer composed of from 20 to 100% by weight of at least one phosphate monomer (a) unit of the following general formula (1), from 0 to 80% by weight of at least one (meth)acrylate monomer (b) unit, and from 0 to 30% by weight of the other

<u>.</u>

٥,

monomer (c) unit copolymerizable with them, and the glass transition temperature of the copolymer is 80°C or less:

$$CH_2=C$$

$$C \longrightarrow C \longrightarrow C$$

$$C \longrightarrow$$

wherein, R¹ in the formula represents hydrogen atom or methyl group, each R² and R³ independently represents hydrogen atom, or an alkyl group or an alkyl ether group having from 1 to 8 carbon atoms, or an aryl groyp, and A represents an alkylene group having from 2 to 14 carbon atoms or a polyoxyalkylene group.

6. (Amended) A raw material composition for preparing a vibration-damping material, which is composed of from 20 to 100% by weight of at least one phosphate monomer (a) of the following general formula (1), from 0 to 80% by weight of at least one (meth)acrylate monomer (b), and from 0 to 30% by weight of the other monomer (c) copolymerizable with them:

$$CH_2=C$$
 $C \longrightarrow C \longrightarrow C \longrightarrow C$
 $C \longrightarrow$

wherein, R¹ in the formula represents hydrogen atom or methyl group, each R² and R³ independently represents hydrogen atom, or an alkyl group or an alkyl ether group having

form 1 to 8 carbon atoms, or an aryl group, and A represents an alkylene group having from 2 to 14 carbon atoms or a polyoxyalkylene group.

REMARKS

Claims 1 and 3-6 are active in the present application. Claim 2 has been cancelled.

Claims 1 and 6 have been amended. Support for amended Claim 1 is found on page 8, lines

20-24. Support for amended Claim 6 is found in Figure 1 and in the specification on page 10, line 15, through page 11, line 19. No new matter is added. An action on the merits and allowance of claims is solicited.

Respectfully submitted, OBLON, SPIVAK, McCLELLAND, MAIER & NEUSTADT, P.C.

Norman F. Oblon Attorney of Record Registration No. 24,618

Daniel J. Pereira, Ph.D. Registration No. 45,518

22850

(703) 413-3000 Fax #: (703)413-2220 DJPER/kst

I:\atty\SUKOS\212638us-pr.wpd

212638US-0PCT

Marked-Up Copy
Serial No:
Amendment Filed on:

IN THE CLAIMS

Please cancel Claim 2.

--1. (Amended) A copolymer composed of from 20 to 100% by weight of at least one phosphate monomer (a) unit of the following general formula (1), from 0 to 80% by weight of at least one (meth)acrylate monomer (b) unit, and from 0 to 30% by weight of the other monomer (c) unit copolymerizable with them, and the glass transition temperature of the copolymer is 80°C or less:

$$CH_2=C$$
 $C - O - A - O - P$
 $O \cap OR^2$
 $O \cap OR^2$

wherein, R¹ in the formula represents hydrogen atom or methyl group, each R² and R³ independently represents hydrogen atom, or an alkyl group or an alkyl ether group having from 1 to 8 carbon atoms, or an aryl groyp, and A represents an alkylene group having from 2 to 14 carbon atoms or a polyoxyalkylene group.

6. (Amended) A <u>raw material</u> composition <u>for preparing a vibration-damping</u> material, which is composed of from 20 to 100% by weight of at least one phosphate monomer (a) of the following general formula (1), from 0 to 80% by weight of at least one

\$.

<u>.</u> -

(meth)acrylate monomer (b), and from 0 to 30% by weight of the other monomer (c) copolymerizable with them:

$$CH_2=C / C O OR^2$$

$$C - O - A - O - P OR^3$$

$$(1)$$

wherein, R¹ in the formula represents hydrogen atom or methyl group, each R² and R³ independently represents hydrogen atom, or an alkyl group or an alkyl ether group having form 1 to 8 carbon atoms, or an aryl group, and A represents an alkylene group having from 2 to 14 carbon atoms or a polyoxyalkylene group.--

JC03 Recircit 7.0 1 7 AUG 2001

DESCRIPTION

COPOLYMER, VIBRATION-DAMPING MATERIAL, VIBRATION-DAMPING PART AND COMPOSITION FOR COPOLYMER

5

10

Technical Field

The present invention relates to a copolymer excellent in vibration-damping property, flame retardancy and transparency, a vibration-damping material and a vibration-damping part composed of this copolymer, and a composition for copolymer.

Background Art

15

20

Conventionally, there are used a lot of vibration-damping polymer materials utilizing polymer viscoelastic behavior for reducing vibration of metals and the like. Since a vibration-damping material is usually used in the state that it is adhered to a hard substrate such as a metal, acrylic resin-based vibration-damping materials excellent in adhesion have been investigated frequently. For example, JP-A No. 4-202586 discloses a cross-linkable copolymer made of 75 to 92% by weight of a monomer necessarily composed of an alkyl acrylate having C8-12 alkyl group and 8 to 25% by weight of a carboxyl group-containing monomer which a polymer obtained by homo-polymerization of this monomer has a glass transition temperature of 50°C or

more.

5

10

15

20

25

Recently, vibration-damping materials are variedly used, and it is required to have various abilities such as transparency, rigidity, impact resistance and weather resistance in addition to conventional vibration-damping property. Particularly, an acrylic resin-based vibration-damping material is often required to have flame retardancy, and it is desired to achieve a vibration-damping materials having both vibration-damping property and flame retardancy.

Usually, the flame retardancy of resin is imparted by blending a flame retarder. In a general method, an organic flame retarder such as phosphorus-based retarders, nitrogen-based retarders and halogen-based retarders, and an inorganic flame retarder such as antimony trioxide and aluminum hydroxide are used as the flame retarder, and these are blended in the resin. However, when a large amount of flame retarder is added for enhancing flame retardancy, problems should occur, for example, that a flame retarder bleeds onto the surface of the resin to deteriorate transparency, and that the other properties such as mechanical strength are remarkably reduced. Further, when a halide is used as the flame retarder, toxicity thereof is problematical. There are various investigation for solving these problems, and for example, JP-A No. 7-82324 discloses a copolymer made of a monomer which can produce

a transparent polymer when polymerized singly, and a monomer having a polymerizable unsaturated double bond containing phosphorus.

However, the copolymer disclosed in JP-A No. 7-82324 is a hard material used mainly for grazing, and the material cannot be applied to uses requiring flame retardancy and vibration-damping property simultaneously.

Disclosure of Invention

10

5

An object of the present invention is to provide a copolymer excellent in vibration-damping property, flame retardancy and transparency, a vibration-damping material and a vibration-damping part composed of this copolymer, and a copolymer composition.

15

The present inventors have intensively studied for attaining the above-mentioned object, and resultantly found that transparency, flame retardancy and vibration-damping property can be imparted by copolymerizing a phosphorus-containing compound having a specific structure with a (meth)acrylate monomer, leading to completion of the present invention.

20

Namely, the present invention is a copolymer composed of from 20 to 100% by weight of at least one phosphate monomer (a) unit of the following general formula (1), from 0 to 80% by weight of at least one (meth)acrylate monomer (b) unit, and from 0 to 30% by weight of the other

25

10

15

20

monomer (c) unit copolymerizable with them:

wherein, R¹ in the formula represents hydrogen atom or methyl group, each R² and R³ independently represents hydrogen atom, or an alkyl group or an alkyl ether group having from 1 to 8 carbon atoms, or an aryl group, and A represents an alkylene group having from 2 to 14 carbon atoms or a polyoxyalkylene group.

Further, the present invention provide a vibrationdamping material and a vibration-damping part composed of the above-mentioned copolymer.

Furthermore, the present invention provide a composition composed of from 20 to 100% by weight of at least one phosphate monomer (a) of the above-mentioned general formula (1), from 0 to 80% by weight of at least one (meth) acrylate monomer (b), and from 0 to 30% by weight of the other monomer (c) copolymerizable with them.

In the present invention, "(meth)acrylate" means "acrylate and/or methacrylate".

Brief Explanation of Drawing

Fig. 1 shows one example of a vibration-damping part composed of a copolymer of the present invention.

10

15

20

25

Best Mode for Carrying Out the Invention

The phosphate monomer (a) constituting the copolymer of the present invention is represented by the abovementioned general formula (1). Specific examples thereof include (2-hydroxyethyl) acrylate acid phosphate, (2hydroxyethyl) methacrylate acid phosphate, (2-hydroxypropyl) acrylate acid phosphate, (2-hydroxypropyl) methacrylate acid phosphate, (3-hydroxypropyl) acrylate acid phosphate, (3-hydroxypropyl) methacrylate acid phosphate, diphenyl-2-acryloyloxyethyl phosphate and diphenyl-2methacryloyloxyethyl phosphate. These may also be used in combination of two or more.

Of them, diphenyl-2-methacryloyloxyethyl phosphate is preferable from the standpoints of copolymerization property and compatibility with other monomers. Namely, it is preferable to use, as at least one phosphate monomer (a), diphenyl-2-methacryloyloxyethyl phosphate singly, or to use this together with the above-mentioned other phosphate monomer. In the explanation of the invention, a polymer composed of 100% by weight of one kind of phosphate monomer (a) unit is also explained expedientially as the copolymer. Namely, the expression "copolymer" may mean "polymer" in some cases.

The copolymer of the present invention contains a phosphate monomer (a) unit in a proportion of 20% by weight or more. When less than 20% by weight, sufficient

. .

3

5

10

15

20

25

vibration-damping property can not be manifested, and close adherence to hard parts made of metals and the like and flame retardancy also deteriorate. This amount is preferably 25% by weight or more, further preferably 30% by weight or more.

In the present invention, disclosed as examples of the (meth)acrylate monomer (b) are methyl (meth)acrylate, ethyl (meth)acrylate, isopropyl (meth)acrylate, butyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, phenyl (meth) acrylate and benzyl (meth)acrylate. These may also be used in combination of two or more.

The copolymer of the present invention contains a (meth)acrylate monomer (b) unit in a proportion of from 0 to 80% by weight.

As the copolymer of the present invention, those composed of a phosphate monomer (a) unit and a (meth)acrylate monomer (b) unit are preferable. From the standpoint of manifestation of the vibration-damping property of the copolymer, the content of the (meth)acrylate monomer (b) unit in the copolymer is preferably 75% by weight or less, further preferably 70% by weight or less.

For manifestation of excellent vibration-damping property of the copolymer of the present invention, it is preferable that the glass transition temperature of the copolymer (hereinafter, the glass transition temperature of a copolymer or polymer is referred to as "Tg", expedi-

10

15

20

25

entially) is lower than the use temperature Tu of the copolymer, and more preferably, the difference of Tu and Tg is 20°C or more. Tg of the copolymer can be lowered by inclusion a monomer unit in the copolymer wherein the monomer gives lower Tg of a polymer obtained by homopolymerization thereof than Tg of a polymer obtained by homopolymerization of a phosphate monomer (a) unit. From such a standpoint, a (meth)acrylate monomer (b) unit is preferably selected. When two or more (meth)acrylate monomer (b) units are used together, it is preferable to select at least one component according to such a standpoint.

In a copolymer composed of these phosphate monomer

(a) unit and (meth)acrylate monomer (b) unit, the content

of the (meth)acrylate monomer (b) unit is preferably 10%

by weight or more, further preferably 20% by weight or

more. Namely, the content of the phosphate monomer (a)

unit is preferably 90% by weight or less, further prefera
bly 80% by weight or less.

The copolymer of the present invention can contain the other monomer (c) unit as a tertiary component in a proportion of from 0 to 30% by weight which does not reduce the properties of the copolymer, in addition to the above-mentioned two component. The other monomer (c) unit is not particularly restricted. For example, unsaturated carboxylic acids such as acrylic acid, methacrylic acid, maleic acid and itaconic acid; acid anhydrides such as

10

15

20

25

maleic anhydride and itaconic anhydride; maleimide derivatives such as N-phenylmaleimide, N-cyclohexylmaleimide and N-t-butylmaleimide; hydroxyl group-containing monomers such as 2-hydroxypropyl acrylate, 2-hydroxyethyl acrylate and 2-hydroxypropyl methacrylate; nitrogen-containing monomers such as (meth)acrylamide, (meth)acrylonitrile, diacetoneacrylamide and dimethylaminoethyl methacrylate; epoxy group-containing monomers such as allyl glycidyl ether, glycidyl acrylate and glycidyl methacrylate; styrene-based monomers such as styrene and α -methylstyrene; and the like can be used.

It is preferable that the copolymer of the present invention is cross-linked, and if necessary, it can contain a cross-linkable monomer unit. Examples of the typical cross-linkable monomer unit include ethylene glycol diacrylate, allyl acrylate, ethylene glycol di(meth)acrylate, allyl (meth)acrylate, divinylbenzene, trimethylol-propane tri(meth)acrylate, 1,3-butylene di(meth)acrylate and hexamethylene diacrylate.

Further, it is preferable that Tg of the copolymer of the present invention is 80°C or less. When Tg is lowered appropriately, vibration-damping property tends to be improved. Tg is more preferably 50°C or less, particularly preferably 20°C or less.

At 25°C as the usual use temperature, it is preferable that $\tan\delta$ of the copolymer of the present invention

10

15

20

25

is 0.5 or more under the condition of 10 hz.

When environmental pollution is taken into consideration, it is preferable that the copolymer of the present invention does not contain a halogen atom.

The composition of the present invention is composed of from 20 to 100% by weight of at least one phosphate monomer (a), from 0 to 80% by weight of at least one (meth)acrylate monomer (b), and from 0 to 30% by weight of the other monomer (c) copolymerizable with them. As the specific examples of these monomers (a) to (c), the abovementioned monomers are listed. Further, the composition can contain the above-mentioned cross-linkable monomer. In the present invention, a material of single component composed of 100% by weight of one phosphate monomer (a) is also referred to as "composition", expedientially.

The viscosity of this composition can be controlled by previously polymerizing a part of the monomer or the monomer mixture, or by adding a suitable high molecular weight polymer or a thickening agent. Further, if necessary, a chain transfer agent, a coloring agent, an ultraviolet absorber, a heat stabilizer, an antistatic agent, a rubbery polymer and fillers such as a inorganic filler can be added.

The copolymer of the present invention can be produced by a known method such as emulsion polymerization, solution polymerization and bulk polymerization. Of them,

10

15

20

25

a bulk polymerization is particularly preferable, and in this case, a copolymer can be produced by using a polymerizable raw material obtained by adding a radical polymerization initiator to the above-mentioned composition, and polymerizing this.

As the radical polymerization initiator, known azo compounds or organic peroxides etc. can be used. As the azo compound, 2,2'-azobisisobutyronitrile, 2,2'-azobis (2,4-dimethylvaleronitrile), 2,2'-azobis(2,4-dimethyl-4-methoxyvaleronitrile) and the like are listed. As the organic peroxide, benzoyl peroxide, lauroyl peroxide, thexyl peroxypivalate and the like are listed. Further, polymerization initiators of redox type, for example, combinations of organic peroxides and amines can also be used.

A polymerizable raw material containing such a radical polymerization initiator added can be, for example, poured between two facing inorganic glass plates or metal plates of which peripheral parts are sealed with a gasket and thermally polymerized to obtain a polymer in the form of a plate, or poured into a part cavity optionally shaped and polymerized under this condition.

The copolymer of the present invention is excellent particularly in vibration-damping of hard parts such as metals, and is suitable as a vibration-damping material. Specifically, the copolymer can be used as a vibration-damping part giving a structure in which the copolymer is

10

15

sandwiched between two hard parts, a structure in which the copolymer is filled in a cavity of a hard part, a structure in which the copolymer is laminated to a hard part, and the like. As the use field of the vibration—damping part, there are listed transportation machine field such as engines, engine covers, bodies of an automobile, tractor and bulldozer; field of domestic electric appliances using a motor such as a washing machine, refrigerator, cleaner and air conditioner; field of OA appliances using a motor such as a personal computer and printer; precise machine filed such as a measuring appliance; construction field of an earthquake damper and piping connection; life field as interiors such as ceiling and wall; and the like.

For example, when a vibration-damping material 2 composed of the copolymer of the present invention is filled between two steel tubes 1 placed concentrically as shown in Fig. 1, excellent vibration-damping property can be obtained.

Next, the present invention will be illustrated further specifically by examples, but the scope of the present invention is not limited by these examples. In the examples, "part" is by weight unless otherwise stated.

Properties in the examples were evaluated by the following methods.

1) Glass transition temperature (Tg)

20

25

3

,

5

Tg of a copolymer resulted was measured by a differential scanning calorimeter DSC200 (manufactured by Seiko Denshi Kogyo K.K.).

2) Vibration-damping property ($\tan \delta$)

A copolymer resulted was cut into a disklike form having a thickness of 3 mm and a diameter of 2.5 cm, and $\tan\delta$ (loss tangent) at 25°C and 10 Hz was measured by using a rheometer RDA-700 (manufactured by Rheometric Scientific FE K.K.).

10 [Example 1]

To a composition composed of a mixture of 53 parts of diphenyl-2-methacryloyloxyethyl phosphate (manufactured by Daihachi Kagaku Kogyo K.K., trade name: MR260) and 47 parts of 2-ethylhexyl acrylate was added 0.2 parts of a polymerization initiator t-hexylperoxypivalate, to give a polymerizable raw material. The polymerizable raw material was subjected to pressure reduction to remove remaining air, then poured into a cell having a thickness of 4.2 mm formed by two stainless plates having a longitudinal size of 460 mm and a transverse size of 610 mm and having inner surfaces to which a Teflon film had been pasted, the peripheral parts thereof being sealed with a gasket. It was polymerized in hot water of 65°C for 120 minutes, and further polymerized under air atmosphere of 100°C for 60 minutes, to obtain a polymer in the form of plate having a thickness of 3 mm. The flame retardancy of this plate

12

polymer was measured according to UL-94 standard, to find it was V-0.

[Example 2]

5

10

15

20

25

The same procedure was conducted as in Example 1 to obtain a plate polymer, except that 53 parts of diphenyl-2-methacryloyloxyethyl phosphate, 27 parts of 2-ethylhexyl acrylate and 20 parts of methyl methacrylate were used as a composition, and 0.065 parts of 2,2'-azobisisobutyronitrile was used as a polymerization initiator.

[Example 3]

The same procedure was conducted as in Example 1 to obtain a plate polymer, except that 43 parts of diphenyl-2-methacryloyloxyethyl phosphate, 47 parts of 2-ethylhexyl acrylate and 10 parts of (2-hydroxyethyl) methacrylate acid phosphate (manufactured by Johoku Kagaku Kogyo K.K., trade name JPA-514) were used as a composition.

[Example 4]

The same procedure was conducted as in Example 1 to obtain a plate polymer, except that 30 parts of diphenyl-2-methacryloyloxyethyl phosphate and 70 parts of butyl acrylate were used as a composition.

[Example 5]

The same procedure was conducted as in Example 1 to obtain a plate polymer, except that 2 parts of benzoyl peroxide (manufactured by Kayaku Akuzo K.K., trade name:

Ž,

J.

5

10

15

20

Kadox B-CH50) and 0.2 parts of N,N'-dimethylaniline were used as a polymerization initiator and polymerization was conducted for 180 minutes under an air atmosphere of 23°C instead of polymerization in hot water of 65°C for 120 minutes.

[Example 6]

The same procedure was conducted as in Example 1 to obtain a plate polymer, except that 0.5 parts of hexamethylene diacrylate was further added as a cross-linking agent to the composition. The resulted polymer was not dissolved in a solvent such as acetone and tetrahydrofuran at all so that it was confirmed that this polymer was a cross-linked body.

Tg and $\tan \delta$ of the plate polymers in Examples 1 to 6 are shown in Table 1.

[Comparative Examples 1 to 3]

The same procedure was conducted as in Example 1 to obtain plate polymers, except that those described in Table 1 were used as a composition, and 0.5 parts of hexamethylene diacrylate was added as a cross-linking agent. Tg and $\tan\delta$ of these plate polymers are shown in Table 1. All of the plate polymers had lower $\tan\delta$ as compared with the plate polymers obtained in Examples, and was not preferable as a vibration-damping material

Table 1

Diphenyl-2- Ex.1 Ex.2 Ex.3 Ex.4 Ex.5 Ex.6 Comp. Gomp. Gomp. Ex.2 methacryloyloxy ethyl phosphate 53 43 30 53 53 0 0 2-Ethylhexyl acrylate 47 27 47 0 47 47 100 80 Methyl methacrylate 0 20 0 0 0 0 0 20 Butyl acrylate 0 <										
Y 53 43 30 53 53 65.1 47 27 47 0 47 47 100 0 20 0 0 0 0 0 1) 0 0 0 0 0 0 1) 0 0 10 0 0 0 1) -35 -5 -39 -30 -35 -30 -80 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0		Ex.1	Ex.2	Ex.3	Ex.4	Ex.5	Ex.6	Comp.	<u> </u>	Comp.
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Diphenyl-2- methacryloyloxy ethyl phosphate	53	53	43	30	53	53	0	EX. 2	Ex. 3
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	2-Ethylhexyl acrylate	47	27	47	0	47	47	100	80	0
1) 0 0 0 70 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	Methyl methacrylate	0	20	0	0	0	0	0	20	0
0 0 10 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	Butyl acrylate	0	0	0	7.0	0	0	0	0	100
o.8 1.2 0.8 0.8 0.8 0.8 0.8 0.2	(2-Hydroxyethyl) methacrylate acid phosphate	0	0	10	0	0	0	0	0	0
0.8 1.2 0.8 0.8 0.8 0.2	Glass transition temperature ($^{\circ}\mathbb{C}$)	-35	5	-39	-30	-35	-30	-80	-55	-50
	tan δ (25 $^{\circ}$ C)	0.8	1.2	0.8	8.0	8.0	0.8	0.2	0.5	0.2

ž

5

Industrial Applicability

The copolymer of the present invention is excellent in balance of transparency, flame retardancy and vibration-damping property, and can be used in various fields such as a vibration-damping material, sound insulation plate, paint, adhesive and various improvers. By polymerizing the composition of the present invention, the abovementioned copolymer can be obtained easily.

CLAIMS

1. A copolymer composed of from 20 to 100% by weight of at least one phosphate monomer (a) unit of the following general formula (1), from 0 to 80% by weight of at least one (meth)acrylate monomer (b) unit, and from 0 to 30% by weight of the other monomer (c) unit copolymerizable with them:

wherein, R^1 in the formula represents hydrogen atom or methyl group, each R^2 and R^3 independently represents hydrogen atom, or an alkyl group or an alkyl ether group having from 1 to 8 carbon atoms, or an aryl group, and A represents an alkylene group having from 2 to 14 carbon atoms or a polyoxyalkylene group.

15

10

5

- 2. The copolymer according to Claim 1 wherein the glass transition temperature is $80\,^{\circ}\text{C}$ or less.
- 3. The copolymer according to Claim 1 wherein $\tan\delta$ at 25°C and 10 Hz is 0.5 or more.
- 4. A vibration-damping material composed of the co-20 polymer of Claim 1.
 - 5. A vibration-damping part composed of the copolymer of Claim 1.
 - 6. A composition composed of from 20 to 100% by

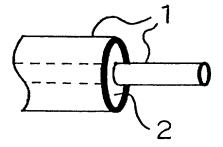
weight of at least one phosphate monomer (a) of the following general formula (1), from 0 to 80% by weight of at least one (meth)acrylate monomer (b), and from 0 to 30% by weight of the other monomer (c) copolymerizable with them:

5

wherein, R^1 in the formula represents hydrogen atom or methyl group, each R^2 and R^3 independently represents hydrogen atom, or an alkyl group or an alkyl ether group having from 1 to 8 carbon atoms, or an aryl group, and A represents an alkylene group having from 2 to 14 carbon atoms or a polyoxyalkylene group.

10

F I G. 1



Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。	As a below named inventor, I hereby declare that:
私の住所、私書箱、国籍は下記の私の氏名の後に記載された通 りです。	My residence, post office address and citizenship are as stated next to my name.
下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者(下記の氏名が一つの場合)もしくは最初かつ共同発明者(下記の名称が数数の場合)であると信じています。	I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled. COPOLYMER, VIBRATION-DAMPING
	MATERIAL, VIBRATION-DAMPING PART AND
1.5 mil.	COMPOSITION FOR COPOLYMER
上記発明の明細書は、 本書に添付されています。	the specification of which is attached hereto. was filed on <u>February 25, 2000</u> as United States Application Number or PCT International Application Number PCT/JP00/01088 and was amended on (if applicable).
私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容 を理解していることをここに表明します。	I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.
私は、連邦規則法典第37編第1条56項に定義されるとおり、特許 資格の有無について重要な情報を開示する義務があることを認 めます。	I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

Japanese Language Declaration

(日本語宣言書)

私は、米国法典第35編119条 (a) - (d) 項又は365条 (b) 項に基づき下記の、米国以外の国の少なくとも一ヵ国を指定している特許協力条約365 (a) 項に基づく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

Prior Foreign Application(s) 外国での先行出願

11-048243	Japan			
(Number)	(Country)			
(番号)	(国名)			
11-215241	Japan			
(Number)	(Country)			
(番号)	(国名)			

私は、第35編米国法典119条 (e) 項に基づいて下記の米国特許 武出願規定に記載された権利をここに主張いたします。

> (Application No.) (出願番号)

(Filing Date) (出願日)

私は、下記の米国法典第35編120条に基づいて下記の米国特許 出願に記載された権利、又は米国を指定している特許協力条約 365条(c)に基づく権利をここに主張します。また、本出願の各 請求範囲の内容が米国法典第35編112条第1項又は特許協力条約で 規定された方法で先行する米国特許出願に開示されていない限 り、その先行米国出願書提出日以降で本出願書の日本国内また は特許協力条約国際提出日までの期間中に入手された、連邦規 則法典第37編1条56項で定義された特許資格の有無に関する重要 な情報について開示義務があることを認識しています。

(Application No.)	(Filing Date)
(出願番号)	(出願日)
(Application No.) (出願番号)	 (Filing Date) (出願日)

私は、私自信の知識に基づいて本宣言書中で私が行なう表明が 真実であり、かつ私の入手した情報と私の信じるところに基づ く表明が全て真実であると信じていること、さらに故意になさ れた虚偽の表明及びそれと同等の行為は米国法典第18編第1001 条に基づき、罰金または拘禁、もしくはその両方により処罰され ること、そしてそのような故意による虚偽の声明を行なえば、 出願した、又は既に許可された特許の有効性が失われることを 認識し、よってここに上記のごとく宣誓を致します。 I hereby claim foreign priority under Title 35, United States Code, Section 119 (a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

	Priority Claimed		
	優先	雀主張	
25/02/1999	X		
(Day/Month/Year Filed) (出願年月日) 29/07/1999	Yes はい 区	No いいえ □	
(Day/Month/Year Filed) (出願年月日)	Yes はい	No いいえ	

I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below.

(Application No.) (Filing Date) (出願番号) (出願日)

I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of application.

(Status: Patented, Pending, Abandoned) (現況:特許許可済、係属中、放棄済)

(Status: Patented, Pending, Abandoned) (現況:特許許可済、係属中、放棄済)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Japanese Language Declaration

(日本語宣言書)

委任状:私は下記の発明者として、本出願に関する一切の手続きを発特許商標局に対して遂行する弁理士または代理人として、 下記の者を指名いたします。

(弁護士、または代理人の指名及び登録番号を明記のこと)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: (list name and registration number)



書類送付先

Send Correspondence to:

022850

直接電話連絡先: (名前及び電話番号)

Direct Telephone Calls to: (name and telephone number)

(703) 413-3000

		
』 ┊ 単独発明者または第一の共同発明 [†]	者の氏名	Full name of sole or first joint inventor Hiroki NAKAMURA
発明者の署名	日付	Inventor's signature Date Hiroki NAKAMURA August 3, 2001
住所		Residence Hiroshima, Japan
国籍		Citizenship Japanese
郵便の宛先		Post Office Address c/o Otake Plants, Mitsubishi Rayon Co., Ltd., 20-1,
		Miyukicho, Otake-shi, Hiroshima 739-0693, Japan
第二の共同発明者の氏名		Full name of second joint inventor, if any Tetsuya SAWANO
第二の共同発明者の署名	日付	Second joint Inventor's signature Date Tetsuya Sawawa August 3, 2001
住所		Residence / Toyama, Japan
国籍		Citizenship Japanese
郵便の宛先		Post Office Address c/o Toyama Plants, Mitsubishi Rayon Co., Ltd.,
		3, Kaigandori, Toyama-shi, Toyama 931-8601, Japan

(第三以降の共同発明者についても同様に記載し、署名すること)

(Supply similar information and signature for third and subsequent joint inventors.)

Page 3 of 3